

AMENDED SPECIFICATION

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PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Improvements in or relating to the manufacture of Polymeric Materials

We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London, S.W.1., a British Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to improvements in or relating to the manufacture of polymeric materials, more particularly polyurethane materials.

It has already been proposed to manufacture polyurethane materials by interacting organic polyisocyanates with hydroxyl group-containing polymers such as polyethers, polyesters of polyesteramides, optionally in the presence of catalysts. It has also been proposed to modify the reaction, for example by the addition of water, in order to produce blown, cellular materials.

It is known to incorporate further ingredients, for example fireproofing agents, in the reaction mixture from which such polyurethane materials are made, but such further ingredients frequently require to be added in such high proportions that other properties of the final product are adversely affected. It is highly desirable that such properties as the fire-resistance of the product should be uniform throughout, but this is not readily achieved when the fire-proofing agent is a solid since such solids are generally insoluble in the polyurethane-forming reagents and it is not easy to secure a satisfactorily even dispersion of the ingredients. Furthermore, the relatively high density of the solid tends to cause even a suitable dispersion to be difficult

to maintain, for example during storage.

We have now found that these difficulties may be overcome, and these adverse effects upon the product may be avoided, by using a hydroxyl-containing polymer which itself contains combined halogen, in conjunction with an organic bromine compound which is soluble in the reaction mixture.

Thus according to our invention we provide an improved process for the manufacture of polyurethane materials by interacting an organic polyisocyanate with a hydroxyl group-containing polymer such as a polyester, polyesteramide or polyether, characterised in that there is used a hydroxyl group-containing polymer which itself contains combined halogen atoms in its structure, in conjunction with an organic bromine compound soluble in one or more of the polyurethane-forming reagents.

The hydroxyl group-containing polymer which itself contains combined halogen may be for example a polyester, polyesteramide or polyether, and may be made by conventional methods provided that at least one of the starting materials from which the hydroxyl group-containing polymer is made is a halogenated compound. Examples of suitable halogen-containing starting materials from which the hydroxyl group-containing polymer may be derived include 2:3:4:5-tetrachlorophthalic acid, hexachloro endomethylene tetrahydrophthalic acid and chloromaleic acid. The corresponding fluoro, bromo or iodo compounds are also suitable, and the acids may be used in the form of their anhydrides or other ester-forming derivatives if desired.

The polyesters or polyesteramides may be

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made from polyhydric alcohols and polycarboxylic acids and, as necessary, polyfunctional aminoalcohols or polyamines. In addition to the halogen-containing compounds mentioned above, suitable polycarboxylic acids include succinic, glutaric, adipic, β -methyl adipic, azelaic, sebacic, isosebacic, phthalic, isophthalic and terephthalic acids or mixtures thereof, and suitable polyhydric alcohols include ethylene, 1:2-propylene, 1:3-propylene, 1:2-butylene, 1:3-butylene, 1:4-butylene, 1:5-pentamethylene, 1:6-hexamethylene, 1:10-decamethylene, diethylene and triethylene glycols, glycerol, hexanetriols and pentaerythritol or mixtures thereof. Examples of polyamides and polyfunctional amines include ethylene, propylene, tetramethylene, pentamethylene, hexamethylene and decamethylene diamines, monoethanolamine, diethanolamine, triethanolamine, polyethylene polyamines, and mixtures thereof.

The polyethers may be, for example, hydroxyl-ended polymers or copolymers of cyclic ethers, and especially of ethylene oxide, propylene oxide, epichlorhydrin, oxacyclobutane and substituted oxacyclobutanes, and tetrahydrofuran. Such polyethers or copolyethers are conveniently prepared by the methods described in U.K. Specification No. 733,624. The halogenated material is most conveniently introduced by using as initiator for the polymerisation of the cyclic ether either the halogenated acid itself or, when a branched polyether is required, the reaction product of the halogenated acid with a polyol, for example the product of reaction of 1 mol. of halogenated diacid with 2 moles of a polyol of functionality 3 or more. Polyfunctional reagents may also be copolymerised with the ether, for example glycidol, glycerol, trimethylolpropane and 3-hydroxymethyloxacyclobutanes.

The quantity of the halogenated starting material used in the preparation of the hydroxyl group-containing polymer should be such that the hydroxyl group-containing polymer contains at least 1%, and preferably at least 5% by weight of combined halogen. In general, larger proportions than 15% may be used if desired but the additional advantage resulting may not be sufficient to justify the extra cost involved. The halogen present is preferably bromine, since thereby the greatest combination of economy and effectiveness of flame-proofing may be achieved, but may be chlorine, iodine or fluorine if so desired.

It is also preferred that the halogen atoms present in the hydroxyl group-containing polymer should be attached to aliphatic carbon atoms, except when such a condition results in the material being unstable under the reaction conditions employed, since thereby the halogen atoms exert their maximum effect in flame-proofing.

The hydroxyl group-containing polymer which itself contains combined halogen may, if desired, be used in admixture with other hydroxyl group-containing polymers which do not contain halogen provided they are mutually soluble in the proportions used.

The organic bromine compound, soluble in one or more of the polyurethane-forming reagents, should contain at least 10%, and preferably at least 30% by weight of bromine. In general, the optimum effect is obtained when the organic bromine compound contains a high proportion of bromine, since thereby the quantity required for a given effect is kept to a minimum. It is usually not practicable, however, to use compounds having a bromine content greater than about 90%. The bromine compound may be isocyanate-reactive, in which case it is resistant to loss by evaporation, migration or extraction by solvents, or may be inert, as desired. It is preferred that the bromine atoms are attached to aliphatic rather than aromatic carbon atoms.

Examples of bromine compounds suitable for dissolving in the hydroxyl group-containing polymer include tris(β -bromoethyl) phosphate, tris(2:3-dibromopropyl) phosphate, brominated paraffin waxes, benzotribromide, bromoform, benzyl bromide, 2-bromoethanol, 2:3-dibromopropanol, 2:3-dibromobutanol, 1:4-dihydroxy-2:3-dibromobutane, 2:2:2-tribromoethanol, 2:3-dibromopropionic acid, 2:3-dibromo-2-methylpropionic acid, methyl-3:4:5:6-tetrabromophthalate, methylhexabromo-endomethylene tetrahydrophthalate and brominated anilines, N-methylanilines, o-toluidines, m-toluidines, p-toluidines, N-methyl-o-toluidines, N-methyl-m-toluidines and N-methyl-p-toluidines.

Examples of bromine compounds suitable for dissolving in water, when this is a reagent in the polyurethane-forming mixture used, as in the preparation of foams, include bromal hydrate.

The fire-retarding properties of the product may be improved still further by the addition of solid arsenic, antimony or bismuth compounds, particularly the oxide or sulphide, but this modification involves the inconvenience resulting from the use of solid additives. Nevertheless, if this inconvenience can be tolerated, an enhanced fire-proofing effect may be obtained. An improved fire-proofing effect may also be obtained by the addition of solid arsenic, antimony or bismuth compounds in the absence of the added brominated flame-proofing material, but again the above objections apply.

The proportion of the organic bromine compound to be employed is usually between 5% and 25%, preferably 10%—20%, of the weight of the hydroxyl group-containing polymer. Larger proportions may be used if desired but little additional advantage is

thereby achieved, and smaller proportions may be used, but the increase in fireproofing effect thereby achieved is usually not sufficient for general utility.

- 5 As examples of polyisocyanates which may be used in the process of the present invention, there may be mentioned tolylene-2:4- and -2:6-diisocyanates, *m*- and *p*-phenylene diisocyanates, chlorophenylene-2:4-diisocyanate, polymerised tolylene-2:4- and 10 -2:6-diisocyanates, 2:4:6-triisocyanato toluene, or mixtures thereof. The polyisocyanates may be partially polymerised or may be modified by treatment with glycols 15 or polyhydric alcohols.

- The polyurethane products may be homogeneous or cellular, rigid or flexible, and may be made by continuous or discontinuous mixing methods, as desired. The invention is 20 especially valuable in the manufacture of foamed, cellular materials, and particularly of rigid products.

- Other known adjuvants may also be added to the reaction mixture from which the polyurethanes are made, for example catalysts, 25 surface active agents, fillers, plasticisers and the like and, in the case of foamed polyurethanes, water as necessary.

- The invention is illustrated but not limited 30 by the following Examples in which the parts and percentages are by weight.

EXAMPLE 1.

- A mixture of 408 parts of pentaerythritol, 945 parts of 1:3-butylene glycol, 1314 parts 35 of adipic acid and 583 parts of hexachloroendomethylene tetrahydrophthalic acid was heated and stirred in a stream of carbon dioxide gas at temperatures up to 160°C.

whilst distilling off water of reaction. The product was a brown, liquid polyester of acid 40 value 33.6 mg. KOH/g., hydroxyl value 221.6 mg. KOH/g. and viscosity of 392 poises at 25°C.

Into 100 parts of this polyester were dispersed by stirring 4 parts of water, 1 part 45 of the condensation product of octyl cresol with 9.5 molecular proportions of ethylene oxide, and 15 parts of 2:3-dibromopropanol. 125 Parts of a diphenylmethane diisocyanate composition, prepared by phosgenating crude 50 diaminodiphenylmethane, containing about 15% of polyamines (mainly triamines) obtained by condensing formaldehyde with aniline in the presence of hydrochloric acid, were then added and the whole was 55 vigorously stirred for 2 minutes. The mixture was then poured into a mould, where it foamed to produce a rigid, cellular structure of fine texture. The above procedure was repeated with the omission of the 2:3- 60 dibromopropanol.

Samples of the resulting foams were tested for flame resistance, using the following test:

A laminate 7" square and 1½" thick is 65 cut from the foam to be tested. The flame from a Bunsen burner at full gas pressure and maximum air inlet is directed at an angle of 45° to the vertical on to the cut face of the laminate, so that the tip of the blue cone of the flame is ¼" from the surface 70 of the foam. After 10 seconds flame application the gas supply is cut off, and then the time for all the flames to be extinguished is measured. This time is taken as a measure 75 of the degree of flame resistance of the foam.

The results of the tests were as follows:—

	Density (lbs. per cubic foot)	Flame Extinction Time (seconds)
Foam containing 2:3-dibromopropanol	2.51	2.5
Foam made in the absence of 2:3-dibromopropanol	2.23	20

EXAMPLE 2.

- 80 A mixture of 181 parts of pentaerythritol, 420 parts of 1:3-butylene glycol, 584 parts of adipic acid, 154 parts of tetrabromophthalic anhydride and 51 parts of phthalic anhydride was heated and stirred in a stream 85 of carbon dioxide gas at temperatures up to 165°C. whilst distilling off water of reaction. The product was a brown liquid polyester of acid value 28.2 mg. KOH/g., hydroxyl value 240 mg. KOH/g. and viscosity of 418 poises at 25°C.

- 90 Into 100 parts of this polyester were dispersed by stirring 4 parts of water, 1 part

of the condensation product of octyl cresol with 9.5 molecular proportions of ethylene oxide, and 15 parts of 2:3-dibromopropanol. 95 120 Parts of the diphenylmethane diisocyanate composition of Example 1 were added, and the whole was vigorously stirred for 2 minutes. The mixture was then poured into a mould where it foamed to produce a 100 rigid cellular structure of fine texture.

The above procedure was repeated with the omission of the 2:3-dibromopropanol.

The densities and flame extinction times, determined by the method described in 105 Example 1, were as follows:—

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	Density (lbs. per cubic foot)	Flame Extinction Time (seconds)
Foam containing 2:3-dibromopropanol	2.66	3.0
Foam made in the absence of 2:3-dibromopropanol	2.66	25

In our earlier Application No. 27038/58 (Serial No. 895,966) we have claimed an improved process for the manufacture of polymeric materials by interacting an organic polyisocyanate with a hydroxyl group-containing material such as a polyester, polyesteramide or polyether, characterised in that there is incorporated therein at least one compound containing a halogen or phosphorous atom and also at least one isocyanate reactive group in the molecule.

WHAT WE CLAIM IS:—

1) Improved process for the manufacture of polyurethane materials by interacting an organic polyisocyanate with a hydroxyl group-containing polymer such as a polyester, polyesteramide or polyether, characterised in that there is used a hydroxyl group-containing polymer which itself contains combined halogen atoms in its structure, in conjunction with an organic bromine compound soluble in one or more of the polyurethane-forming reagents.

2) Improved process for the manufacture of polyurethane materials as claimed in claim 1 wherein the quantity of the halogenated starting material used in the preparation of the hydroxyl group-containing polymer is such that the hydroxyl group-containing polymer contains at least 1%, and preferably at least 5% by weight of combined halogen.

3) Improved process for the manufacture of polyurethane materials as claimed in claim 1 or claim 2 wherein the halogen in the hydroxyl group-containing polymer is bromine.

4) Improved process for the manufacture of polyurethane materials as claimed in any

of the preceding claims wherein the halogen atoms in the hydroxyl group-containing polymer are attached to aliphatic carbon atoms.

5) Improved process for the manufacture of polyurethane materials as claimed in any of the preceding claims wherein the organic bromine compound contains at least 10%, and preferably at least 30% by weight of bromine.

6) Improved process for the manufacture of polyurethane materials as claimed in any of the preceding claims wherein the bromine atoms in the organic bromine compound are attached to aliphatic carbon atoms

7) Improved process for the manufacture of polyurethane materials as claimed in any of the preceding claims wherein the proportion of the organic bromine compound employed is between 5% and 25%, preferably between 10% and 20%, of the weight of the hydroxyl group-containing polymer.

8) Improved process for the manufacture of polyurethane materials as claimed in any of the preceding claims when applied to the manufacture of foamed, cellular materials, and particularly of rigid products.

9) Improved process for the manufacture of polyurethane materials substantially as hereinbefore described especially with reference to the foregoing Examples.

10) Polyurethane materials whenever prepared by a process claimed in any of the preceding claims or by an obvious chemical equivalent thereof.

WALTER SCOTT,
Agent for the Applicants.

PROVISIONAL SPECIFICATION

Improvements in or relating to the manufacture of Polymeric Materials

We, IMPERIAL CHEMICAL INDUSTRIES LIMITED of Imperial Chemical House, Millbank, London, S.W.1, a British Company do hereby declare this invention to be described in the following statement:—

This invention relates to improvements in or relating to the manufacture of polymeric materials, more particularly polyurethane materials.

It has already been proposed to manufacture polymeric materials by interacting organic polyisocyanates with hydroxyl group-containing materials such as polyethers, polyesters or polyesteramides, optionally in the presence of catalysts. It has also been proposed to modify the reaction, for example by the addition of water, in order to produce blown, cellular materials.

It is known to incorporate further ingredients, for example fireproofing agents, in the reaction mixture from which such polymeric materials are made, but such further ingredients frequently require to be added in such high proportions that other properties of the final product are adversely affected. It is highly desirable that such properties as the fire-resistance of the product should be uniform throughout, but this is not readily achieved when the fire-proofing agent is a solid since such solids are generally insoluble in the polyurethane-forming reagents and it is not easy to secure a satisfactory even dispersion of the ingredients. Furthermore, the relatively high density of the solid tends to cause even a suitable dispersion to be difficult to maintain, for example during storage.

We have now found that these difficulties may be overcome, and these adverse effects upon the product may be avoided, by using a hydroxyl-containing material which itself contains combined halogen, in conjunction with another organic halogen compound which is soluble in the reaction mixture.

Thus according to our invention we provide an improved process for the manufacture of polymeric materials by interacting an organic polyisocyanate with a hydroxyl group-containing material such as a polyester, polyesteramide or polyether, characterised in that there is used a hydroxyl group-containing material which itself contains combined halogen atoms in its structure, in conjunction with an organic halogen compound soluble in one or more of the polyurethane-forming reagents.

The hydroxyl group-containing material which itself contains combined halogen may be for example a polyester, polyesteramide or polyester, and may be made by conventional methods provided that at least one of the starting materials from which the hydroxyl group-containing material is made is a halogenated compound. Examples of suitable halogen-containing starting materials from which the hydroxyl group-containing material may be derived include 2:3:4:5-tetrachlorophthalic acid, hexachloroendomethylene tetrahydrophthalic acid and chloromaleic acid. The corresponding fluoro, bromo or iodo compounds are also suitable, and the acids may be used in the form of their anhydrides or other ester-forming derivatives if desired.

The polyesters or polyesteramides may be made from polyhydric alcohols and polycarboxylic acids and, as necessary, polyfunctional aminoalcohols or polyamines. In addition to the halogen-containing compounds mentioned above, suitable polycarboxylic acids include succinic, glutaric, adipic, β -methyl adipic, azelaic, sebacic, isosebacic, phthalic, isophthalic and tere-

phthalic acids or mixtures thereof, and suitable polyhydric alcohols include ethylene, 1:2-propylene, 1:3-propylene, 1:2-butylene, 1:3-butylene, 1:4-butylene, 1:5-pentamethylene, 1:6-hexamethylene, 1:10-decamethylene, diethylene and triethylene glycols, glycerol hexanetriols and pentaerythritol or mixtures thereof. Examples of polyamines and polyfunctional amines include ethylene, propylene, tetramethylene, pentamethylene, hexamethylene and decamethylene diamines, monoethanolamine, diethanolamine, triethanolamine, polyethylene polyamines, and mixtures thereof.

The polyethers may be, for example, hydroxyl-ended polymers or copolymers of cyclic ethers, and especially of ethylene oxide, propylene oxide, epichlorhydrin, oxacyclobutane and substituted oxacyclobutanes, and tetrahydrofuran. Such polyethers or copolyethers are conveniently prepared by the methods described in U.K. Specification No. 733,624. The halogenated material is most conveniently introduced by using as initiator for the polymerisation of the cyclic ether either the halogenated acid itself or, when a branched polyether is required, the reaction product of the halogenated acid with a polyol, for example the product of reaction of 1 mol. of halogenated diacid with 2 moles of a polyol of functionality 3 or more. Polyfunctional reagents may also be copolymerised with the ether, for example glycidol, glycerol, trimethylolpropane and 3-hydroxymethyloxacyclobutanes.

The quantity of the halogenated starting material used in the preparation of the hydroxyl group-containing material should be such that the hydroxyl group-containing material contains at least 1%, and preferably at least 5% by weight of combined halogen. In general, larger proportions than 15% may be used if desired but the additional advantage resulting may not be sufficient to justify the extra cost involved. The halogen present is preferably bromine, since thereby the greatest combination of economy and effectiveness of flame-proofing may be achieved, but may be chlorine, iodine or fluorine if so desired.

It is also preferred that the halogen atoms present in the hydroxyl group-containing material should be attached to aliphatic carbon atoms, except when such a condition results in the material being unstable under the reaction conditions employed, since thereby the halogen atoms exert their maximum effect in flame-proofing.

The hydroxyl group-containing material which itself contains combined halogen may, if desired, be used in admixture with other hydroxyl group-containing materials which do not contain halogen provided they are mutually soluble in the proportions used.

The organic halogen compound, soluble in

one or more of the polyurethane-forming reagents, should contain at least 10%, and preferably at least 30% by weight of halogen.

In this instance also, it is preferred that the halogen should be bromine though it may be chlorine, iodine or fluorine if desired. In general, the optimum effect is obtained when the organic halogen compound contains a high proportion of halogen, since thereby the quantity required for a given effect is kept to a minimum. It is usually not practicable, however, to use compounds having a halogen content greater than about 90%. The halogen compound may be isocyanate-reactive, in which case it is resistant to loss by evaporation, migration or extraction by solvents, or may be inert, as desired. It is preferred that the halogen atoms are attached to aliphatic rather than aromatic carbon atoms.

Examples of halogen compounds suitable for dissolving in the hydroxyl group-containing material include tri-(β -chloroethyl)phosphate, tri-(2:3-dichloropropyl)phosphate, chlorinated paraffin waxes, benzotrichloride, chloroform, benzyl chloride, 2-chloroethanol, 2:3-dichloropropanol, 2:3-dichlorobutanol, 1:4-dihydroxy-2:3-dichlorobutane, 2:2:2-trichloroethanol, 2:3-dichloropropionic acid, 2:3-dichloro-2-methylpropionic acid, methyl-3:4:5:6-tetrachlorophthalate, methyl-hexachloro-*endo*-methylene tetrahydrophthalate, and chlorinated anilines, N-methylanilines, *o*-toluidines, *m*-toluidines, *p*-toluidines, N-methyl-*o*-toluidines, N-methyl-*m*-toluidines and N-methyl-*p*-toluidines. There may also be mentioned the corresponding fluoro, bromo and iodo compounds.

Examples of halogen compounds suitable for dissolving in water, when this is a reagent in the polyurethane-forming mixture used, as in the preparation of foams, are chloral hydrate and the corresponding fluoro, bromo and iodo compounds.

The fire-retarding properties of the product may be improved still further by the addition of solid arsenic, antimony or bismuth compounds, particularly the oxide or sulphide, but this modification involves the inconvenience resulting from the use of solid additives. Nevertheless, if this inconvenience can be tolerated, an enhanced fire-proofing effect may be obtained. An improved fire-proofing effect may also be obtained by the addition of solid arsenic, antimony or bismuth compounds in the absence of the added halogenated flame-proofing material, but again the above objections apply.

The proportion of the organic halogen compound to be employed is usually between 5% and 25%, preferably 10%—20%, of the weight of the hydroxyl group-containing material. Larger proportions may be used if desired but little additional advantage is

thereby achieved, and smaller proportions may be used, but the increase in fire-proofing effect thereby achieved is usually not sufficient for general utility.

As examples of polyisocyanates which may be used in the process of the present invention, there may be mentioned tolylene-2:4- and -2:6-diisocyanates, *m*- and *p*-phenylene diisocyanates, chlorophenylene-2:4-diisocyanate, polymerised tolylene-2:4- and -2:6-diisocyanates, 2:4:6-triisocyanato toluene, or mixtures thereof. The polyisocyanates may be partially polymerised or may be modified by treatment with glycols or polyhydric alcohols.

The polyurethane products may be homogeneous or cellular, rigid or flexible, and may be made by continuous or discontinuous mixing methods, as desired. The invention is especially valuable in the manufacture of foamed, cellular materials, and particularly of rigid products.

Other known adjuvants may also be added to the reaction mixture from which the polyurethanes are made, for example catalysts, surface active agents, fillers, plasticisers and the like and, in the case of foamed polyurethanes, water as necessary.

The invention is illustrated but not limited by the following Examples in which the parts and percentages are by weight.

EXAMPLE 1.

A mixture of 408 parts of pentaerythritol, 945 parts of 1:3-butylene glycol, 1314 parts of adipic acid and 583 parts of hexachloro-*endo*-methylene tetrahydrophthalic acid was heated and stirred in a stream of carbon dioxide gas at temperatures up to 160°C. whilst distilling off water of reaction. The product was a brown, liquid polyester of acid value 33.6 mg. KOH/g., hydroxyl value 221.6 mg. KOH/g. and viscosity of 392 poises at 25°C.

Into 100 parts of this polyester were dispersed by stirring 4 parts of water, 1 part of condensation product of octyl cresol with 9.5 molecular proportions of ethylene oxide, and 15 parts of 2:3-dibromopropanol. Parts of a diphenylmethane diisocyanate composition, prepared by phosgenating crude diaminodiphenylmethane, containing about 15% of polyamines (mainly triamines) obtained by condensing formaldehyde with aniline in the presence of hydrochloric acid, were then added and the whole was vigorously stirred for 2 minutes. The mixture was then poured into a mould, where it foamed to produce a rigid, cellular structure of fine texture. The above procedure was repeated using 15 parts of tri-(β -chloroethyl)phosphate in place of the 15 parts of 2:3-dibromopropanol, and again repeated with the omission of the 2:3-dibromopropanol.

Samples of the resulting foams were tested for flame resistance, using the following test:

- 5 A laminate 7" square and $1\frac{1}{2}$ " thick is cut from the foam to be tested. The flame from a Bunsen burner at full gas pressure and maximum air inlet is directed at an angle of 45° to the vertical on to the cut face of the laminate, so that the tip of the

blue cone of the flame is $\frac{1}{4}$ " from the surface of the foam. After 10 seconds flame application the gas supply is cut off, and then the time for all the flames to be extinguished is measured. This time is taken as a measure of the degree of flame resistance of the foam. 15

The results of the tests were as follows:—

	Density (lbs. per cubic foot)	Flame Extinction Time (seconds)
Foam containing 2:3-dibromopropanol	2.51	2.5
Foam containing tri-(β -chloroethyl)-phosphate	2.53	2
Foam containing neither of the above ingredients	2.23	20

EXAMPLE 2.

- 20 A mixture of 181 parts of pentaerythritol, 420 parts of 1:3-butylene glycol, 584 parts of adipic acid, 154 parts of tetrabromophthalic anhydride and 51 parts of phthalic anhydride was heated and stirred in a stream of carbon dioxide gas at temperatures up to 165°C . whilst distilling off water of reaction. 25 The product was a brown liquid polyester of acid value 28.2 mg. KOH/g., hydroxyl value 240 mg. KOH/g. and viscosity of 418 poises at 25°C . 30

Into 100 parts of this polyester were dispersed by stirring 4 parts of water, 1 part of the condensation product of octyl cresol with 9.5 molecular proportions of ethylene

oxide, and 15 parts of 2:3-dibromopropanol. 35 120 Parts of the diphenyl-methane diisocyanate composition of Example 1 were added, and the whole was vigorously stirred for 2 minutes. The mixture was then poured into a mould where it foamed to produce a rigid cellular structure of fine texture. 40

The above procedure was repeated using 15 parts of tri-(β -chloroethyl)phosphate in place of the 15 parts of 2:3-dibromopropanol, and again repeated with the omission of the 2:3-dibromopropanol. 45

The densities and flame extinction times, determined by the method described in Example 1, were as follows:—

	Density (lbs. per cubic foot)	Flame Extinction Time (Seconds)
Foam containing 2:3-dibromopropanol	2.66	3.0
Foam containing tri(β -chloroethyl)-phosphate	2.80	2.5
Foam containing neither of the above ingredients	2.66	25

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